

Isolated sub-100-attosecond pulse generation via controlling electron dynamics

Pengfei Lan, Peixiang Lu*, Wei Cao, Yuhua Li, Xinlin Wang

*Wuhan National Laboratory for Optoelectronics and School of Optoelectronics Science and Engineering,
Huazhong University of Science and Technology, Wuhan 430074, P. R. China*

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A new method to coherently control the electron dynamics is proposed using a few-cycle laser pulse in combination with a controlling field. It is shown that this method not only broadens the attosecond pulse bandwidth, but also reduces the chirp, then an isolated 80-as pulse is straightforwardly obtained and even shorter pulse is achievable by increasing the intensity of the controlling field. Such ultrashort pulses allow one to investigate ultrafast electronic processes which have never been achieved before. In addition, the few-cycle synthesized pulse is expected to manipulate a wide range of laser-atom interactions.

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In the past decade, there has been a great interest to extend the pulse duration to the attosecond (as) domain. This is sprung by the great potential of attosecond pulses to trace the electronic dynamics in atoms and molecules [1, 2, 3]. The straightforward attosecond metrology prefers an isolated attosecond pulse rather than a train of attosecond pulses [3, 4]. Hence much effort is paid to produce the isolated pulse [3, 4, 5]. It has been shown that an isolated attosecond pulse can be generated with a few-cycle laser pulse by selecting the continuous harmonics in the cutoff [2, 3, 5]. The intractable problem is that the bandwidth of the continuous harmonics in the cutoff is less than 20 eV, and thus the minimum duration of the isolated attosecond pulse is ~ 250 as [2]. This is greater than the characteristic timescale of the electronic process in atoms (152 as, i.e., the period for electrons in the Bohr orbit of ground-state hydrogens), and then the application of the 250-as pulse is significantly limited. Therefore, it is urgently desired to produce an isolated attosecond pulse with broader bandwidth and shorter duration [6, 7, 8, 9]. It is theoretically demonstrated that broadband continuous harmonics can be produced using a few-cycle laser pulse with a modulated polarization [7]. This scheme has been implemented recently [8, 9], and broadband continuous harmonics are observed. However, there is a chirp in these harmonics, then an isolated 280-as pulse is produced straightforwardly. With the technology of chirp compensation [9], the pulse duration is reduced to 130 as. In this letter, we propose a new facile method for isolated sub-100-attosecond pulse generation via controlling electron dynamics using a few-cycle laser pulse in combination with a controlling field.

High harmonic generation (HHG) is well understood in terms of the semiclassical “three-step” model [10]. In detail, the electron is first ionized from the atom, then it oscillates almost freely in the laser field and gains kinetic energy E_k , finally, it recombines with the parent ion by releasing an energetic photon of $I_p + E_k$ where I_p is the ionization

* Corresponding author: lupeixiang@mail.hust.edu.cn

energy. One can control the HHG by manipulating different steps. It has been shown that the synthesized field is an efficient method for coherent controlling electron dynamics (the second step) [11, 12, 13]. In the two-color laser pulse, the harmonic plateau can be extended to higher energy [12, 13]. In addition, the symmetry of the driving field in adjacent half-cycle is broken [11], then the electron motion is different in neighboring half-cycles [12, 13], which allows one to produce an isolated attosecond pulse using a multi-cycle laser pulse of 24-fs in combination with a weak sub-harmonic controlling field, or a 10-fs pulse in combination with a second-harmonic controlling field [14]. However, the bandwidth of the attosecond pulse is only about 10 eV, the pulse duration is greater than 200 as [13, 14]. Unlike these works, we aim to produce broadband supercontinues harmonics and an isolated broadband attosecond pulse with a duration approaching 1 atomic units via coherently controlling the electron dynamics using an ultrashort laser pulse in combination with a controlling field. It is shown that the ultrashort synthesized laser pulse not only broadens the harmonic bandwidth, but also reduces the harmonic chirp, then an isolated 80-as pulse is straightforwardly obtained. By increasing the intensity of the controlling field, the bandwidth can be further broadened, an isolated attosecond pulse of 65 as is straightforwardly generated. Using the technology of chirp compensation as in Ref. [9], the pulse duration is expected to be reduced to about 1 atomic unit of time, allowing one to investigate ultrafast electronic processes which have never been achieved before.

To demonstrate our scheme, we first investigate the HHG process with the semiclassical model [10], which gives us a clear physics picture. The electric field is given by $\mathbf{E}(t) = E_0 f(t) \cos[\omega_0(t - T/2) + \phi] \hat{\mathbf{x}} + E_1 f(t) \cos[\omega_1(t - T/2)] \hat{\mathbf{x}}$. $\hat{\mathbf{x}}$ is the polarization vector, E_0 and E_1 are the amplitudes, ω_0 and ω_1 are the frequencies of the driving and controlling fields, respectively, ϕ is the relative phase. $f(t) = \sin^2(\pi t/T)$ is the pulse envelope, the time t evolves from 0 to T . Fig. 1(a) illustrates the scheme of HHG from neon driven by a few-cycle laser pulse. The laser intensity is 8.3×10^{14} W/cm², $T = 5T_0$ where $T_0 = 2.7$ fs is the period of the driving laser pulse, and the pulse duration is about 5 fs full width at half maximum. Fig. 1(b) shows the dependence of the kinetic energy E_k on the ionization (\bullet) and recombination times (\times). As shown in Fig. 1(a), the 5-fs pulse contains only two optical cycles, thus the electron is only dominantly ionized near the peak, and then three dominant returns are present. For the first return (R_1), the electron is ionized at $t = 1.5T_0$, and returns to the parent nucleus at about $t = 2.2T_0$ with a maximum kinetic energy of 140 eV, emitting harmonic photons with the maximum energy of $I_p + 140$ eV. The maximum energies of the harmonics generated in the second and third returns (R_2 and R_3) become $I_p + 160$ and $I_p + 130$ eV, respectively. Therefore, only the second return (R_2) contributes to the harmonics greater than $I_p + 140$ eV. An isolated attosecond pulse can be obtained by filtering the highest harmonics. However, the bandwidth of this attosecond pulse is about 20 eV, and the minimum pulse duration is about 250 as [2].

By adding a controlling field (with a different frequency) to the few-cycle driving field, the electron dynamics can be modulated. Fig. 2 illustrates the sketch of this method. The dotted line in Fig. 2(a) shows the 5-fs driving laser pulse, the dashed line shows the controlling field with a frequency of $2\omega_0$, and the solid line is the synthesized field. The intensity of the controlling field is 4% of the driving field. By adjusting the relative phase, the controlling field can be set in the same direction with the driving field in the half cycle of $t = 2.5T_0$ (the return R_2), then the electron will gain much higher energy since the driving field is enhanced. In the adjacent half cycles (the returns R_1 and R_3), the

driving and controlling fields change their directions and are in opposite directions, then driving field is weakened and the electron gains less energy. This larger contrast between the neighboring half-cycles will broaden the bandwidth of the continuous harmonics in the cutoff, which leads to an isolated broadband attosecond pulse. Note that similar results can also be achieved by adjusting the delay between the two laser pulses. Fig. 2(b) shows the dependence of the kinetic energy E_k on the ionization (\bullet) and recombination times (\times) in the synthesized field. In contrast to Fig. 1(b), one can see from Fig. 2(b) that the maximum kinetic energy gained in the return R_2 is increased to 180 eV, and those of R_1 and R_3 are decreased to 100 and 120 eV, respectively. Therefore, an isolated attosecond pulse can be produced by superposing all the harmonics greater than $I_p + 120$ eV, i.e., the bandwidth of the attosecond pulse is broadened up to 60 eV, corresponding to an isolated attosecond pulse of about 70 as in the Fourier transform limit. Note that there are many degrees of freedom for choosing the controlling field. Fig. 2(c) and (d) show the HHG in the 10-fs driving laser pulse in combination with a sub-harmonic controlling field. The intensity of the driving laser pulse is 5×10^{14} W/cm², and the controlling field is 4% of the driving field. One can see that the maximum kinetic energies of the three dominant returns are 90, 160, 85 eV, respectively. Then an isolated attosecond pulse with a bandwidth of 70 eV will be generated, which corresponds to an isolated 50-as pulse in the Fourier transform limit. It is worth noting that these results are obtained using a 10-fs laser pulse with an intensity of 5×10^{14} W/cm² in the later case. In contrast to the 5-fs pulse shown in Fig. 2(a), the 10-fs laser system is relatively easier achieved, and the needed intensity is lower.

Following, we investigate the harmonic and attosecond pulse generation with a quantum model [15]. For the calculation, the full electric field of the laser pulse is used, i.e., the nonadiabatic effect is taken account [15]. Further, the neutral depletion is also considered by the ADK ionization rate [15]. Fig. 3(a) shows the harmonic spectrum (bold line) using a synthesized field of a 10-fs driving field and a sub-harmonic controlling field. For comparison, the harmonic spectrum in the 5-fs driving pulse alone is shown by the thin line. One can clearly see that the harmonic spectra show the similar structure. It is chaotic for the low harmonics and becomes regular and continuous for the highest harmonics. It is because many returns contribute to the low harmonics and the interference of these returns leads to a chaotic structure. For the highest harmonics, only one return (R_2) contributes to the HHG, hence a regular and continuous structure is present. Selecting the continuous harmonics, an isolated attosecond pulse is produced. However, in the 5-fs pulse alone, the bandwidth of the continuous harmonics is only about 20 eV. While in the 10-fs synthesized field, the bandwidth of the continuous harmonics is significantly broadened up to 75 eV, an isolated 50-as pulse will be generated in the Fourier transform limit. Further, one can see that there is a regular modulation for the continuous harmonics. This structure can be illustrated with the semiclassical model. As shown in Fig. 1 and 2, there are two couples of ionization and recombination times corresponding the the same kinetic energy in each return. This corresponds to the short and long trajectories, respectively, which also can be retrieved from the quantum model. Fig. 3(b) shows the temporal profiles of the attosecond pulses generated by the harmonics at different central frequencies. As shown in this figure, two attosecond bursts are produced, which are originated from the short and long trajectories, respectively. In terms of Feynman's path-integral theory [16], the electron wave packet takes different quantum paths from the initial state (the atomic ground state) to the final state (the atomic ground state). The phases accumulated

in short and long trajectories are different. Then the interference of these two quantum pathes gives rise to an evident modulation of the harmonic spectrum. The spatial analogy of this phenomena is the Young's two-slit experiment. As shown in Fig. 3(b), the time separation between the short and long trajectories decreases with increasing the harmonic order, then the modulation period increases gradually [see Fig. 3(a)].

It is shown in Fig. 3(b) that the harmonics with different frequencies are emitted at different times. In other words, an intrinsic chirp is present in these harmonics, which is of great importance both from a fundamental point of view as well as for the attosecond pulse generation. To address this critical issue, the emission times of the harmonics calculated with the semiclassical (solid line) and quantum model (open circles) are present in Fig. 4(a). As shown in this figure, the emission time of the short trajectory increases with increasing the harmonic energy, that of the long trajectory decreases, and the emission time approaches to a constant for the harmonics beyond $120\omega_0$. This result demonstrates that the short trajectory has a positive chirp and the long trajectory has a negative chirp. As in the previous work [17, 18], we calculated the chirp rate by $C = \Delta t / \Delta E$ where t and E are the emission time and energy of the harmonics. In an intense driving field [17], the chirp rate is about 9 as/eV , the minimum attosecond pulse imposed by the intrinsic chirp is obtained by selecting the harmonics with a bandwidth of 34 eV , resulting in close-to-Fourier-limited pulses of 130 as . In our scheme, the electron dynamics is significantly modulated by the synthesized field, then the chirp rate is reduced to 5.2 as/eV . By selecting the harmonics with a bandwidth of 54 eV , a close-to-Fourier-limited pulses of 80-as will be generated as shown in Fig. 4(b). Note that the above results are retrieved with a simulation of numerically solving the time-dependent schrödinger equation.

To generate an isolated attosecond pulse, one of the short or long trajectories must be eliminated. It has been shown that the long trajectory leads to a spatially divergent radiation [19]. Therefore by adding a small aperture after the HHG cell, the harmonics and attosecond pulse originated from the long trajectory will be removed. This method has been shown to be efficient in previous studies [20]. Alternatively, it has been demonstrated that the short and long trajectories have much different phase-match conditions [21], and the short trajectory can be selected by focusing the laser pulse before the gas jet. This method has also been implemented and verified experimentally [1, 17]. Taking into account the above discussions, the possible experimental implement of our method is as following. The driving field is straightforwardly produced with a Ti-sapphire laser, the controlling field is produced with down-conversion. Focusing the synthesized field on the gas jet filled with neon, high harmonics will be generated. The laser field and the harmonics bellow $80\omega_0$ can be efficiently absorbed by a Sn foil, then two attosecond bursts with the duration of 80 as will be generated [see Fig. 4(b)]. By adjusting the laser focus or putting a small aperture after the HHG cell, an isolated attosecond pulse will be selected (i.e., the dashed-line pulse in Fig. 4(b) will be removed). It is worth noting that the Sn foil has a negative chirp group delay dispersion. Hence the harmonic chirp will be complemented [17], then an isolated 50-as pulse can be generated by selecting the harmonics with a bandwidth of 75 eV [7].

The isolated 80-as allows one to trace a wide range of ultrafast electronic dynamics which has never been achieved before. Moreover, due to the high photon energy, one can put the insight deeper into the dynamics of inner-shell electrons or even to the nuclear dynamics. In addition, the 80-as pulse contains only 2.9 optical cycles of the central frequency (150 eV). Analogically to the few-cycle infrared pulses [5], such few-cycle isolated attosecond pulses may

pave the way to investigate and manipulate the electronic dynamics when processes are triggered by the electric field of the attosecond pulse rather than by the intensity profile in the extreme ultraviolet regime.

In the above calculation, the intensity of the controlling field is 4% of the driving laser pulse. Our calculation shows that by increasing the intensity of the controlling field, the bandwidth of the attosecond pulse will be further broadened, and the chirp will be further reduced. When the intensity of the controlling field is increased to about 25% of the driving laser pulse, the bandwidth of the continuous harmonics will be increased to 180 eV, and an isolated attosecond pulse of 65 as can be obtained straightforwardly. After compensating the chirp, an attosecond pulse of about 25 as will be produced in the Fourier transform limit. Our calculation indicates that in our scheme the duration of the controlling pulse is flexible, which can be increased to 20 fs, whereas the duration of driving laser pulse plays a vital role. To achieve the broadband attosecond pulse, the driving pulse must be less than 7 fs in the second harmonic controlling field case (Fig. 2(a)), and must be less than 15 fs in the sub-harmonic controlling field case (Fig. 2(c)). If a longer pulse is used, the nonadiabatic effect becomes weak, and the supercontinuum bandwidth will significantly decrease. Then, alike previous works [13, 14], it only supports to an isolated attosecond pulse greater than 200 as. While it should be noted that these laser conditions are presently available, facilitating an easy demonstration of our scheme.

In summary, we propose a new method for coherently controlling the electron dynamics using a few-cycle laser pulse in combination with a controlling field. It is shown that this method can broaden the bandwidth and reduce the chirp of the attosecond pulse simultaneously, then an isolated 80-attosecond pulse is straightforwardly obtained. By increasing the intensity of the controlling field, the bandwidth can be significantly broadened up to 180 eV, an isolated attosecond pulse of 65 as is straightforwardly generated. After compensating the chirp, the pulse duration is expected to be reduced to about 1 atomic unit of time. Such an ultrashort pulse allows one to investigate ultrafast electronic processes which have never been achieved before and to manipulate the electronic dynamics upon changing the pulse phase in the extreme ultraviolet regime. In addition, the few-cycle synthesized pulse allows one to manipulate the electron dynamics more powerfully, which may be used to control the other laser-atom interaction processes.

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FIG. 1: (color online) (a) A sketch of electron dynamics in a 5-fs laser pulse alone. (b) The dependence of kinetic energy of electron on the ionization (\bullet) and recombination times (\times). The laser intensity and wavelength are $8.3 \times 10^{14} \text{ W/cm}^2$ and 800 nm, respectively.

FIG. 2: (color online) (a) The electric fields of the synthesized field (solid line) of a 5-fs driving laser pulse (dotted line) in combination with its second-harmonic controlling pulse (dashed line). The intensity of the controlling field is only 4% of the driving field and the relative phase $\phi = 0.2\pi$. (b) The dependence of kinetic energy of electron on the ionization (\bullet) and recombination times (\times) in the synthesized field shown in (a). (c) and (d) are the same with (a) and (b), but a sub-harmonic controlling field is adopt. The intensity and duration of the driving field are $5 \times 10^{14} \text{ W/cm}^2$ and 10 fs and relative phase $\phi = 0$.

FIG. 3: (color online) (a) The spectra of the HHG in the 10-fs synthesized field with the same parameters of Fig. 2(c) (red bold line) and 5-fs field alone with the same parameters of Fig. 1 (blue thin line). (b) Temporal profiles of the attosecond pulses generated by harmonics with different central frequencies. The bandwidth of the selected harmonics is 30 eV.

FIG. 4: (color online) (a) The emission time of the harmonics calculated with the classical model (solid line) and quantum model (open circles). (b) The temporal profiles of the attosecond pulse by selected the harmonics with a bandwidth of 54 eV ($80\omega_0$ to $115\omega_0$). The solid and dashed lines correspond to the contribution from the short and long trajectories, respectively.

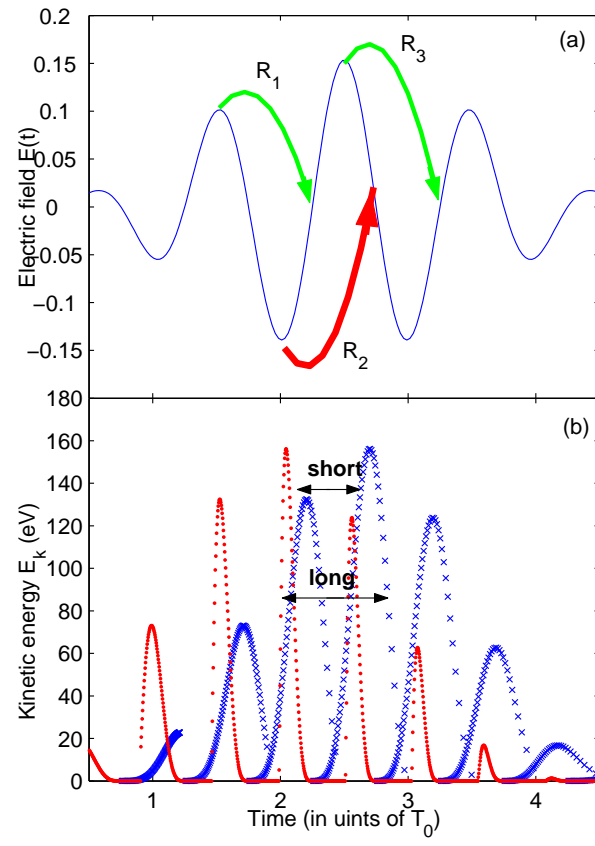


Fig. 1, Lan *et al*

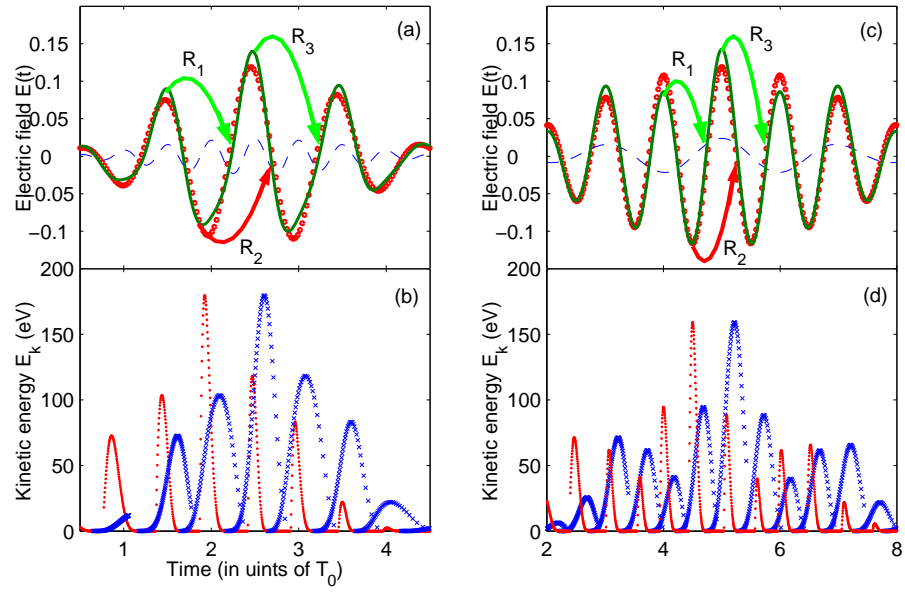
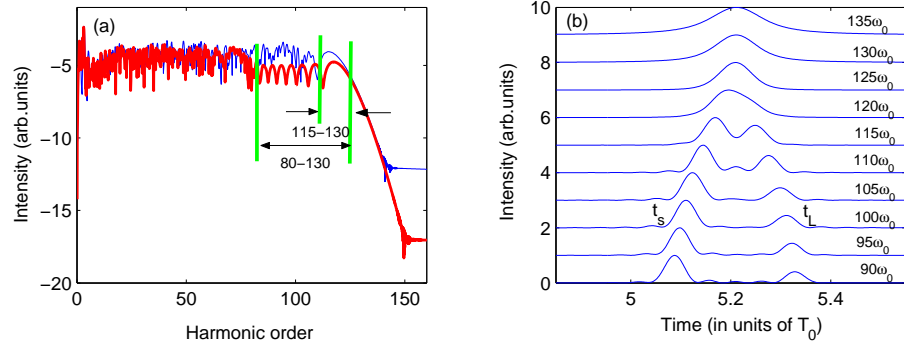
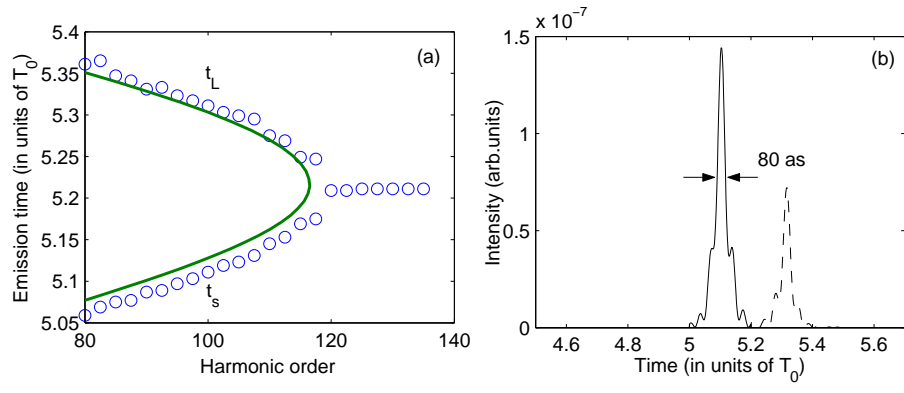


Fig. 2, Lan *et al*

Fig. 3, Lan *et al*Fig. 4, Lan *et al*